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EVALUATION OF PLUTONIUM AT ENEWETAK ATOLL

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Abstract-An extensive survey was carried out in 1972-1973 to assess the current radiological status of Enewetak Atoll. The radionuclides detected in the Atoll environment were studied for their potential contributions to the dose commitment of the returning population according to several pathways of exposure. Plutonium was detected in air and in the terrestrial and aquatic environment at concentrations that varied from background levels due to world-wide fallout to levels several orders-of-magnitude above. The dose commitments from plutonium via the terrestrial food chain and inhalation vary according to the postulated living pattern. The dosages via marine foods can be expected to be insensitive to living pattern and to exceed those via terrestrial foods. Plutonium would contribute nearly all of the dosage via inhalation, but this pathway ranks low in overall importance compared with the food-chain and external-dose pathways. Although the potential dose from plutonium via all pathways is low relative to that from ^{60}Co, ^{90}Sr and ^{137}Cs, plutonium will still remain in the Atoll environment after the other major isotopes have decayed away.

INTRODUCTION

ENEWETAK Atoll in the central Pacific Ocean 2380 naut. miles southwest of Honolulu was the site of 43 nuclear weapons tests carried out by the U.S. from 1948 to 1958. Testing resulted in the deposition of fission products, activation products, and nuclear fuel materials in the terrestrial and aquatic environment of the Atoll, in particular in the north and northeast portions.

In late 1972 the AEC carried out a radiological survey of Enewetak aimed at gathering data needed to develop clean-up and rehabilitation procedures for the resettlement of the Enewetak people to their homeland. An intensive measurements and evaluation program was carried out to define the present distribution of radionuclides and their significance to the radiation exposures of an indigenous population. This paper is drawn from the results of the survey (NVO, 1973) and highlights the findings with special emphasis upon results obtained for plutonium.†

Enewetak Atoll consists of 40 islands on an elliptical coralline reef, approx 23 x 17 naut. miles, with the long axis running northwest to southeast (Fig. 1). The total land area is but 2.75 miles^2, and the mean height above sea level is only 10 ft. The large, central, ocean lagoon is 388 miles^2 in area.

The Atoll climate is tropical marine with high humidity and about 60 in/yr of precipitation. The islands are vegetated heavily with native and introduced species. The Enewetak people have not inhabited the Atoll since testing, so cultivation has been absent and the system appears to be succeeding toward a forest of native trees. Succession has been arrested in some areas as a result of continuing military operations on the Atoll which involved construction and earth moving. While the terrestrial ecosystem is relatively simple, the marine ecosystem is complex and biologically active, typical of tropical reef and lagoon communities.

The Enewetak people, returning, can be expected to practice nonintensive agriculture for coconuts, pandanus, breadfruit and arrowroot and to rely heavily upon the abundant

* This work was performed under the auspices of the U.S. Atomic Energy Commission.

† Unless otherwise stated, plutonium refers to measured ^{239}Pu and ^{240}Pu together. ^{238}Pu, ^{240}Pu and ^{241}Pu are present in the Atoll, but constitute less than 10% of the total plutonium. ^{241}Am is also present and is still increasing slightly due to decay of ^{241}Pu; however, dose contributions expected from ^{241}Am are less than those from Pu.

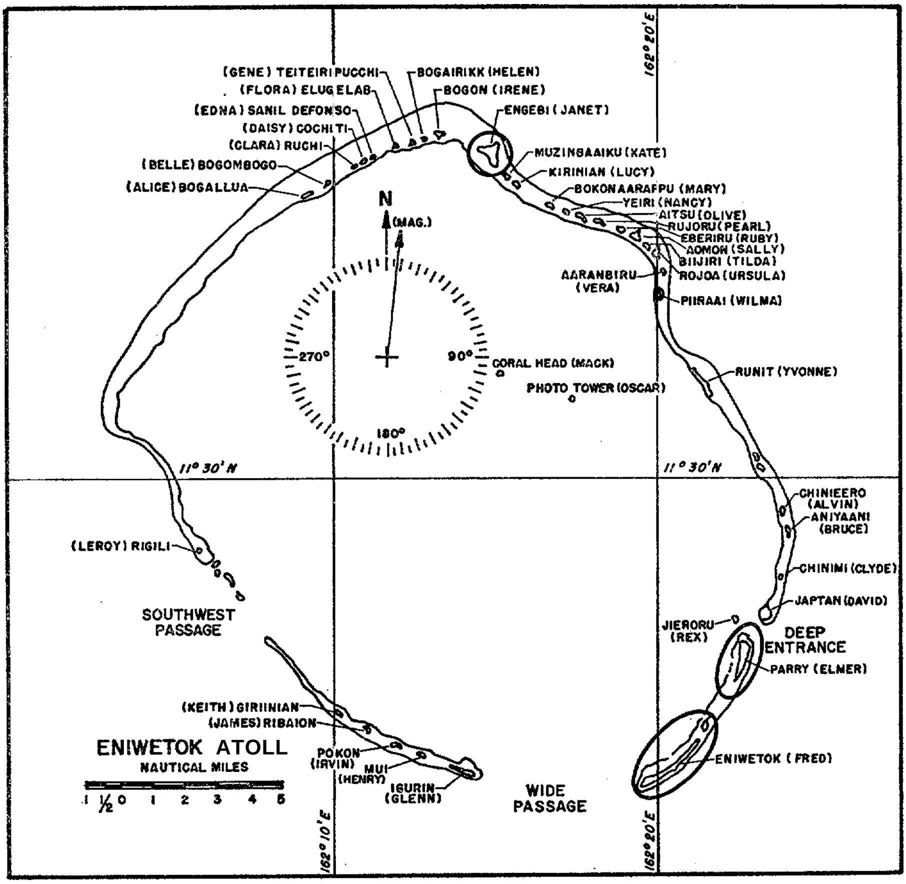

FIG. 1. Enewetak Atoll with true island names and corresponding U.S. site names. Encircled islands are the islands of choice for residence by the Enewetak people.

marine resources of the Atoll. They will continue to import rice, flour, sugar and other staple foods. Resettlement will involve a major program in replanting desired species for food and cash crops such as coconuts. While the population of over 400 people will choose to live centrally on one or more of the large islands, they will utilize most all of the Atoll for agricultural and food gathering purposes. Village areas will be chosen, and houses, community structures, and roads will be built. The choices among the various options in living patterns (agriculture practices, villages, houses, dietary habits) are important variables in the radiological assessment.

HIGHLIGHTS OF THE SURVEY METHODS

The survey was a detailed documentation of present radioactivity levels in soil, sediments, water, air and biota. The sampling design was developed to meet the needs for assessing all the potentially significant pathways of dose to man. In the terrestrial environment, measurements of radioactivity were made of the soil and biota of all islands, so that assessments could be made island-by-island. To the extent possible, edible crops were sampled, but the low numbers of edible plants limited the data base. As an alternative, abundant and ubiquitous species of nonedible vegetation were measured to obtain an understanding of the distribution

Table 1. Plutonium in Enewetak Soils, pCi/g

Island	In Top 15 cm Median	Range	In Top 2 cm Median	Range
Alice	12	3.9-68	56	3.9-105
Belle			96	12-230
Dense	26	7.2-130		
Sparse	11	5.8-26		
Clara	22	3.8-88	40	11-80
Daisy			50	8-180
Dense	41	22-98		
sparse	15	3.8-33		
Edna	18	13-24	18	
Irene	11	2.4-280	13	2.3-43
Janet	8.5	0.08-170	21	2.8-100
Kate			28	1.8-62
Dense	17	8.6-50		
sparse	2.3	0.17-14		
Lucy	7.7	2.4-22	34	8.0-49
Mary	8.0	2.0-35	18	2.0-26
Nancy	9.1	2.3-28	23	9.6-35
Percy	3.5	1.5-23	11	5.5-16
Olive			54	2.8-87
Dense	7.7	2.2-30		
sparse	2.8	1.9-4.1		
Pearl			70	4.0-400
Hot Spot	51	15-530		
Remainder	11	0.85-100		
Ruby	7.3	3.0-24	2.7	
Sally	4.3	0.21-130	18	1.7-62
Tilda			5.8	2.0-16
Dense	7.6	1.4-17		
sparse	2.5	1.1-34		
Ursula	1.3	0.26-7.3	1.5	0.6-2.7
Vera	2.5	0.60-25	22	1.5-35
Wilma	1.1	0.1-5.3	3.3	1.2-7.0
Southern Yvonne	3.2	0.02-50	10	0.24-32
Northern Beaches	2.7	0.34-18		
David, Elmer, Fred	0.04	0.004-0.31	0.12	0.01-0.90
Leroy	0.63	0.02-2.0	1.7	1.1-2.6
All Other Southern Islands	0.07	0.004-1.1	0.12	0.01-0.45

of radionuclides in vegetation on each island. Thorough study was made of the gamma ray fields produced by ^{137}Cs and ^{60}Co in soils.

Similarly, a large number of samples were obtained from the water and sediments of the marine ecosystem, resulting in a detailed inventory of radionuclides in the ecosystem. Marine biota were extensively sampled with consideration for the preferred diet of the Enewetak people.

Air sampling was carried out over the ocean and on several islands to measure the amount of radioactivity in the air due to resuspended particles from the soil. Several kinds of air samplers were employed to determine levels close to heavily contaminated areas, to examine particle size ranges in the suspended aerosol, and to look for evidence of any systematic elevation of radioactivity in air in the vicinity of the islands.

Necessarily, not much detail of the survey techniques nor the resulting data can be reproduced here. Readers desiring more detail are directed to the survey report (NVO, 1973).

HIGHLIGHTS OF SURVEY RESULTS ON PLUTONIUM

The plutonium distribution in Enewetak soils is variable over the Atoll, with levels in the southern islands corresponding to values expected due to world-wide fallout, to small areas containing hundreds of pCi of plutonium/g of soil on northern islands (Table 1). The measured distribution is consistent with that expected on the basis of testing history.

The amount of plutonium in soil generally decreases with depth down the soil column, but the effects of more recent earth moving in some locations are noticeable in inverting the depth profile. The general trend can be seen by comparing the median values for plutonium in the top 2 cm of soil with values for the top 15 cm of soil. A small area on northern Yvonne contains the highest levels of plutonium measured and an unpredictable depth distribution.

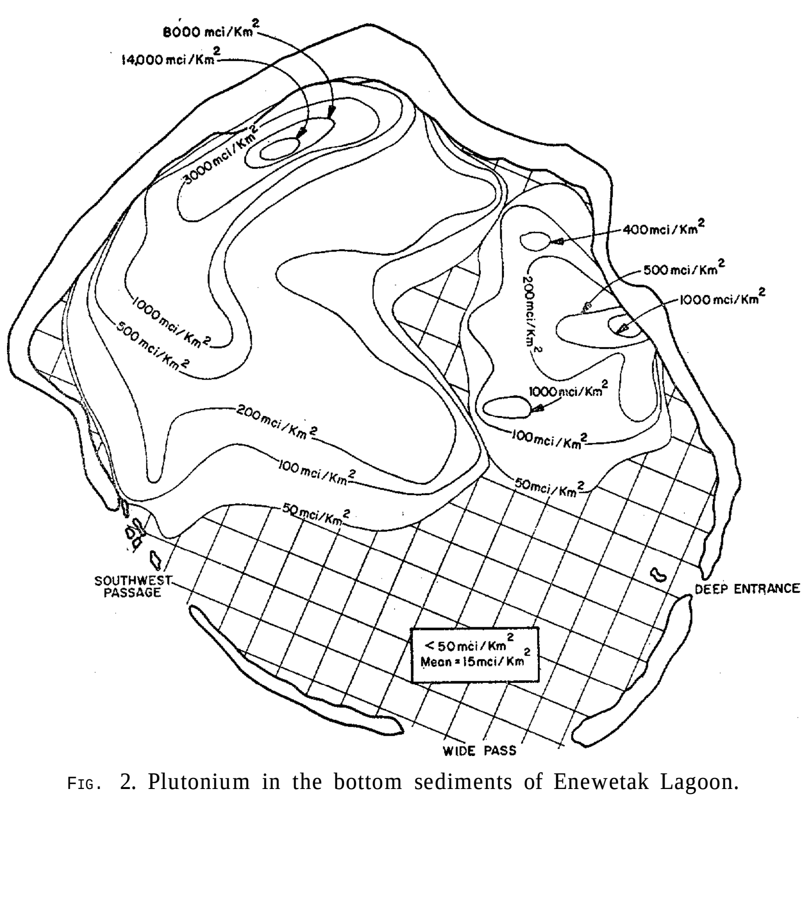

FIG. 2. Plutonium in the bottom sediments of Enewetak Lagoon.

In this area, in spite of the appearance of some samples containing as high as 3000 pCi/g, most samples taken contained less than 100 pCi/g.

The largest inventory of plutonium at Enewetak was measured in the marine bottom sediments. Plutonium is non-umformally distributed over the lagoon floor with highest levels in the northeast (see Fig. 2). The mean plutonium concentration in bottom sediments of the lagoon was 463 mCi/km^2, some 200 times world-wide fallout background. Crater areas in the lagoon near Alice and Belle were the highest plutonium areas measured on the Atoll.

The plutonium concentration in lagoon water ranged from 43 fCi/l. in the northeast quadrant to 9 fCi/l. in the southeast quadrant. The ocean east of Enewetak contained 0.3 fCi/l. Surface waters in the lagoon on the average contained 30% more plutonium than did deep water, suggesting that runoff from the reef and land is a more important source of plutonium to the lagoon water than the large burden of plutonium in the bottom sediments.

The surface atmosphere of the Atoll contained plutonium in a range of <0.003-0.008 fCi/m^3, which is comparable to levels expected due to world-wide fallout. Extreme conditions of high winds led to levels of <0.04-0.13 fCi/m^3 downwind from a plutonium hot spot on Yvonne. While survey results showed that air levels were very low, it is conceivable that human activities, say in village areas would be a sufficient disturbance of soil surfaces to result in higher air loading with plutonium.

The major emphasis in the marine food chain study was on sampling of the reef fish which will comprise the largest diet component from the marine ecosystem. Almost 900 marine specimens were obtained for radionuclide analysis. The specimens were pooled and analyzed in four categories to determine any differences between species or location of catch. The concentration of plutonium in fish was not strongly dependent on the species or on the location of the catch. The average concentration for pelagic fishes plus reef fishes (mullet, goatfish and convict surgeon) was 0.248 pCi/g, range 0.005 to 23 pCi/g (see Table 2).

Over 1000 specimens of plants, birds, eggs, rats and land crabs were taken in the terrestrial biota survey (see Table 3). Coconut was obtained on 16 islands, but only a few samples of pandanus and tacca (arrowroot), and no breadfruit were obtained. *Messerschmidia* and *Scaevola,* and other indicator plants were obtained throughout the Atoll. Birds, bird eggs and rats were plentiful for sampling, but coconut crabs were taken only from a few of the southern islands.

The greatest concentration of plutonium in coconut was 0.036 pCi/g in a sample from Irene. The concentrations in coconut from the other

Table 2. Summary of plutonium concentrations in muscle of fish at Enewetak

Species	No. of Samples	Concentration (pCi/g dry wt) Average a	Average b	Standard Deviation	Range	Lognormal Median
Mullet	25	0.981	0.984	4.60	0.000482 - 23.1	0.0145
Surgeon	28	0.0768	0.0772	0.169	0.00428 - 0.887	0.0260
Goatfish	21	0.0123	0.0130	0.0151	0.00161 - 0.0531	0.00778
Other Fish	49	0.0637	0.0700	0.238	0.000786 - 1.21	0.00909
All Fish	123	0.244	0.248	2.08	0.000482 - 23.1	0.0126

[a]If a measurement was below the limits of detection, the concentration was set equal to zero.

[b]If a measurement was below the limits of detection, the concentration was set equal to the limits of detection.

Table 3. Plutonium in terrestrial biota at Enewetak Atoll

Type of Sample	Concentrations (pCi/g dry wt) Min(Island)	Max(Island)
Coconut		
Meat	0.00013(Vera)	0.0362(Irene)
Milk	a	
Pandanus		
Fruit	0.0034(Belle)[b]	
Leaves	0.0013(David)	0.015(Sally)
Tacca		
Corm	0.0011(David)[b]	
Birds		
Muscle	0.0010(Keith)	0.12(David)
Liver	0.0015(Janet)	0.062(Alice)
Eggs	0.00045(Yvonne)	0.015(Janet, Sally)
Coconut Crab		
Muscle	0.00076(James)	0.0031(Leroy)
Hepatopancreas	0.0019(James)	0.0098(Keith)
Messerschmidia	0.0019(Irene)	0.77(Yvonne)
Scaevola	0.00050(David)	1.29(Yvonne)
Rats		
Viscera	0.0074(Elmer)	1.77(Pearl)
Muscle	0.0023(Janet)	0.12(Pearl)
Liver	0.008(Sally, Yvonne)	0.041(Ursula)
Bone	0.030(Janet)	3.25(Janet)

[a]Measurements at levels below the limits of detection.
[b]Only a single sample was collected.

islands were less by an order-of-magnitude or more or were below the limits of detection. The only concentration reported in pandanus fruit, 0.0034 pCi/g, was for the sample from Belle. Concentrations in pandanus leaves varied from 0.0013 pCi/g in the sample from David to 0.015 pCi/g in that from Sally. The concentration in the single sample of tacca corm (David) was 0.0011 pCi/g. Concentrations in muscle and liver of birds varied irregularly from island to island and ranged from 0.0010 to 0.12 pCi/g in muscle and from 0.0015 to 0.062 pCi/g in liver. The range- of plutonium concentrations in bird eggs was 0.00045-0.015 pCi/g. The range in muscle of coconut crabs was 0.00076-0.0031 pCi/g, that in hepatopancreas was 0.0019-0.0098 pCi/g. The concentrations in *Messerschmidia* and *Scaevola* from the same location were comparable and varied over an extreme range of 0.00050 pCi/g on David to 1.29 pCi/g on Yvonne. Plutonium levels in rat tissues were greatest in bone and decreased in the order bone, viscera, liver and muscle. The greatest concentration in bone, 3.25 pCi/g, was measured in a rat taken from Janet.

EVALUATION OF PATHWAYS

Exposures to plutonium were evaluated for the inhalation of airborne dust, and for doses derived from ingestion of foods from terrestrial and marine food chains. Calculations were made on an island-by-island basis using the survey measurements data obtained for each island. These results were applied to a hypothetical living pattern model which had the island of residence and islands used for agriculture as variables. Two of the living patterns are covered here. These cases are solutions which include the likely islands of residence, Fred and Janet (see Fig. 1):

Living Pattern I: Live on Fred and Elmer, carry out agriculture on Alvin through Keith

Living Pattern III: Live on Janet and carry out agriculture on Janet.

In both living patterns, fishing is carried out over the entire Atoll. These two living patterns approximate a limiting low radiation back. ground case (in the southern islands) and **a** limiting high radiation background case (living and farming in the north only). Other living patterns can be constructed and evaluated by using the results of the island-by-island assessment.

Inhalation pathway

It has been well documented that radioactivity in soils can resuspend in the atmosphere and be available for inhalation. Quantitative models of this pathway and the doses obtainable from a given level of contamination are under development, but there are large uncertainties inherent in them. These limitations are discussed in detail in NVO-140 and in a paper by Anspaugh *et al.* (1975) in this issue of *Health Physics.* A completely empirical approach to assessing this pathway would require a large measurement program carried out under actual conditions of occupation of the Atoll. As an alternative, a predictive model was developed which utilizes world-wide data on dust loading in the atmosphere, the soil measurements at Enewetak, and a set of conservative assumptiona discussed below.

It is important to provide an evaluation that considers, as far as possible, the potential for exposure to a returned population, which takes into account both the population distribution on the Atoll and the patterns of living. Under conditions of habitation, large areas of soil surface will become stabilized by cultivated vegetation, coral layering in the village areas, and by buildings. These activities will tend to reduce the possibility for resuspension of soil particles. However, human activities such as construction, earth moving, agricultural activities, and children playing, tend to stir up dust. Exposure levels to individuals will depend on such local sources.

The objectives of the measurements and this assessment were to obtain sufficient understanding to place this pathway for dosage in perspective to the food chain pathways. The air sampling program showed that resuspension levels were measurably above background only in the downwind vicinity of scattered hot spots of soil contamination.

Estimates of potential population exposures were made by applying the model,

$$Dt = C_s \cdot Rt \cdot L_a$$

where :

Dt = dose in rem to organ of reference after t yr of continued exposure

C_s = concentration of plutonium in soil, pCi/g

Rt = dose conversion factor, rem/pCi/m^3, for cumulative dose to organ of reference from t yr of continued exposure

L_a = soil loading in air, g/m^3.

This model assumes that plutonium concentrations in soil are conserved in the resuspension process. A value of 100 μg/m^3 was used for L_a, based upon evaluation of a set of measurements of dust in non-urban U.S. air (NAPCA, 1968). In this study, mean dust loads ranged from 9 to 79 μg/m^3 with an overall average of about 40 μg/m^3. Enewetak soils are moist and dust loading is minimal; however, a value of 100 μg/m^3 was adopted for L_a to insure a reasonably conservative accounting for the close proximity of individuals to sources of soil disturbance.

Calculations were carried out for each island using soil measurements of plutonium (see Table 1), and dose conversion factors derived by BENNETT (1974a) using the ICRP Lung Dynamics Model for 0.4-μm plutonium fallout particles of low solubility. Calculations were made for the O-2 cm of soil samples and for the O-l 5 cm samples.

Table 4. Cumulative rems in 70 yr to organs from plutonium via inhalation pathway (based upon plutonium concentration in 0-2 cm of roil)

Living Pattern	Lung	Liver	Bone
I (southern Islands)	0.002	0.002	0.004
III (Live an Janet)	0.306	0.289	0.612

Results of these calculations (see Table 4) show that the population exposures to be expected fall in the mrem range for a lifetime of occupation. Occupation of the southern islands, Living Pattern I, results in organ doses in 70 yr the order of a few mrem, while occupation of northern islands will lead to organ doses approaching a rem in 70 yr. The highest dosages from this pathway were calculated for occupation of Belle, 2.77 rem in 70 yr. These results indicate that the inhalation pathway will not be a significant feature of population dosages. Removal of hot spot radioactivity and land use planning can insure that plutonium resuspension is a negligible pathway to man in the rehabilitated Atoll.

Terrestrial and marine food chains

Two dietary cases were developed (see Table 5) ; the first, which is associated with the time of return is distinguished by the absence of pandanus fruit, breadfruit and arrowroot. These items appear only in the 10-yr post-return diet since it requires several years for these crops to reestablish and come to full production. This

Table 5. Postulated diet for the returning adult Enewetak population

	Diet, g/day	
Food item	At time Of return	10 yr post-return
Fish	600	600
Domestic meat	60	100
Pandanus fruit	0	200
Breadfruit	0	150
Wild birds	100	20
Bird eggs	20	10
Arrowroot	0	40
Coconut	100	100
Coconut milk	100	300
Coconut crabs	25	25
Clams	25	25
Imports	200-1000	2004000
	1030 plus imports	1570 plus imports

diet is based on the carefully considered judgement of persons who have lived in the Marshall Islands in intimate association with the native residents and therefore have first-hand knowledge of their living habits. Accurate assessment of the role of imported foods is not possible, but it is generally recognized that imported foods, such as rice, flour, tea, canned fish and canned meats, will be major components of the diet. Accordingly, the dosage via food chains may be overestimated to the extent that imported food items will replace food items listed in the table.

Many of the items expected in the terrestrial diet were not available in sufficient quantity for adequate sampling at the time of the survey, and other constituents of the ecosystem, i.e. soil and native plants and animals were sampled. In the case of plutonium, prediction of the concentrations in terrestrial foods was carried out following two basic approaches. In one approach it was based largely on the concentrations measured in edible or indicator species. Pandanus leaves served as the direct indicator for pandanus fruit, breadfruit and coconut, and rats served as the direct indicator for domestic meat (pork and chicken) wherever edible species were not sampled or the measurements were inadequate. The concentration of plutonium in a given edible or indicator item for a particular island group was determined as the mean of the samples collected in that island group. The concentration in meat was determined as the overall mean in muscle and liver of rats. In the other approach, prediction was based largely on concentrations estimated from data on average soil concentration and concentration factors between indicator species and soil. Pandanus leaves and rats served in similar fashion as indicator species. The two approaches are not entirely independent, since in each case the concentrations of plutonium in birds, bird eggs and coconut crabs were based on measurements.

Concentration factors were determined by pairing plant or animal and soil data for the same location and then calculating pCi/g dry plant or animal tissue per pCi/g soil (see Tables 6 and 7). The concentration in meat was calculated assuming a concentration factor of 0.005, the overall median for muscle and liver of rat (see Table 6). The concentration in pandanus leaves was estimated using a concentration factor of 0.003, the median concentration factor for *Messerschmidia* and *Scaevola* (see Table 7). The average concentration of plutonium in the O-l 5-cm soil profile was computed from Table 1 to be 0.065 pCi/g for island group Alvin-Keith and 8.5 pCi/g for Janet.

It is not surprising that the ranges of concentration factor vary greatly by as much as factors of 10^3 or more since this procedure is inherently lacking in precision. However, the major fraction of the calculated concentration factors is within an order-of-magnitude of the median value. Although the concentrations of

Table 6. Relationship between plutonium concentrations in animal tissues and soil

Species, Tissue	No.	Concentration Factor (pCi/g dry tissue ÷ pCi/g dry soil) Min.	Median	Max
Rat				
Muscle	11	0.00009	0.033[a]	0.79
Liver	13	0.00014	0.0033[a]	2.0
Bone	15	0.0017	0.014	9.1
Pooled	39	0.00009	0.094	9.1
Coconut Crab				
Muscle	5	0.0032	0.013	0.018
Hepatopancreas	4	0.0039	0.026	0.12
Exoskeleton	5	0.0084	0.026	0.037
Pooled	14	0.0032	0.016	0.12
Hermit Crab				
Muscle	6	0.0030	0.093	0.55
Hepatopancreas	4	0.0087	0.051	0.11
Exoskeleton	5	0.00074	0.010	0.18
Pooled	15	0.00074	0.048	0.55
Pooled Coconut Crab & Hermit Crab	29	0.00074	0.026	0.55

[a] Overall median of muscle and liver combined is 0.005.

Table 7. Soil-&-plant uptake of plutonium

Plant Type	No. of Samples	Concentration Factor (pCi/g dry plant ÷ pCi/g dry soil) Min	Median	Max
Messerschmidia	24	0.00017	0.0030	0.20
Scaevola	24	0.00013	0.0031	0.14
Pooled Messerschmidia & Scaevola	48	0.00013	0.0030	0.20
Coconut Meat	2	0.0013		0.0077
Pandanus Leaves	4	0.00046	0.0056	0.01

plutonium in rat tissues were not significantly correlated with those in soil. there were statistically significant correlations between the logarithms of the plutonium concentrations in *Messerschmidia,* Scaevola, and pooled *Messerschmidia* and Scaevola and those in soil *(Messerschmidia P* = 0.05, *Scaevola P* = 0.01, pooled *Messerschmidia* and *Scaevola P* = 0.001). Furthermore the linear regression lines for *Messerschmidia* and Scaevola were found to be statistically indistinguishable with common slopes and common intercepts.

The concentrations of plutonium predicted for terrestrial foods from island groups Alvin–Keith and Janet were selected by the approach yielding the greater values of concentration (see Table 8). Those listed for Alvin-Keith were based on measurements, those for Janet were based primarily on concentration factors and levels in soil, The concentrations are listed according to two different start dates. Meat, birds, bird eggs, coconut crabs and coconut from the southern islands are assumed to be available relatively soon following return to the Atoll and their concentrations are listed under 1 January 1974, the postulated date of return. The edible plants are generally assumed to become available for the first time 8 yr after return and their concentrations are listed under 1 January 1982. Most of the plutonium concentrations are considerably in excess of the concentrations reported in terrestrial foods from the 1972 New York diet, which range from 9×10^{-7} pCi/g fresh weight in canned vegetables to 8.5×10^{-6} pCi/g fresh weight in bakery products (BENNETT, 1974b).

Table 8. Concentrations of plutonium in terrestrial foods

	Concentration, pCi/g dry wt	
	Jan. 1, 1974	Jan. 1, 1982
Island Group Alvin-Keith[a]		
Domestic Meat	0.020	0.020
Pandanus Fruit	n.a.[b]	0.0039
Breadfruit	n.a.	0.0039[c]
Wild Birds	0.0235	0.0235
Bird Eggs	0.00065	0.00065
Arrowroot[b]	n.a.	b
Coconut Meat	0.0016	0.0016
Coconut Milk	0.016[d]	0.016[d]
Coconut Crabs	0.0027	0.0027
Island Group Janet		
Domestic Meat	0.043	0.043
Pandanus Fruit	n.a.	0.026
Breadfruit	n.a.	0.026[c]
Wild Birds	0.0033	0.0033
Bird Eggs	0.0148	0.0148
Arrowroot[b]	n.a.	b
Coconut Meat	n.a.	0.026[c]
Coconut Milk	n.a.	0.26[d]
Coconut Crabs	n.a.	n.a.

[a]The concentration for Alvin-Keith are based on measurements; those for Janet are based primarily on concentration factors and levels in soil.

[b]The abbreviation n.a. signifies not available. Although arrowroot can be expected in the diet beginning 1982, it can be expected to contribute relatively very little plutonium to the terrestrial diet and no concentration has been listed.

[c]The concentration is assumed to be that in pandanus leaves.

[d]The dry wt concentration in coconut milk is assumed to be 10 times that in coconut meat.

Plutonium in the marine food chain was evaluated in four main fish groups-surgeonfish, goatfish, mullet and "other" fish-where "other" includes not only other species of fish but also tridacna clams and lingusta. The mean concentrations in the four fish groups did not differ significantly, and furthermore, dependence of concentrations on island location was not discernible. Accordingly the mean concentration of plutonium in muscle of all fish from the entire Atoll was used to predict exposure to plutonium from the ingestion of marine foods. The overall mean concentration of 0.248 pCi/g was used in the assessment of dose rather than the lognormal median value, which is about a factor of 20 less (see Table 2).

By way of comparison the mean plutonium concentration in fish collected in contaminated waters at Thule, Greenland in 1968 was 0.14 pCi/g fresh weight; the median and mean in soft tissues of bivalves from Zone I were 8.0 pCi/g and 150 pCi/g fresh weight (AARKROG, 1971). The concentration reported for shellfish in the 1972 New York diet was 1.1×10^{-5} pCi/g fresh weight (BENNETT, 1974b). Thus the mean plutonium concentration in fish from Enewetak is comparable to that in fish from Thule and exceeds the concentration reported for shellfish in the 1972 New York diet by more than three orders-of-magnitude.

Rates of ingestion of plutonium were determined by combining the results of Tables 5 and 8. Except for coconut and arrowroot the diets listed in Table 5 refer to ingestion of fresh food. The following values of water content were used to calculate rates of ingestion of plutonium via foodstuffs : coconut meat 50%, coconut milk 95%, pandanus 80%, breadfruit 70%, bird muscle and liver 70%, bird eggs 75%, coconut crab liver 81%, coconut crab hepatopancreas 62%, rat muscle and liver 73% and fish 71% (NVO, 1973). Rates of ingestion (see Table 9) have been computed for the diet at the time of

Table 9. Rate of ingestion of plutonium from terrestrial and aquatic foods

	Ingestion Rate, pCi/day		
	Assuming Diet at Time of Return	Assuming 10-yr Postreturn Diet	
	Jan. 1, 1974	Jan. 1, 1974	Jan. 1, 1982
Island Group Alvin-Keith			
Fish	44	44	
Domestic Meat	0.32	0.54	
Pandanus Fruit			0.16
Breadfruit			0.18
Wild Birds	0.70	0.14	
Bird Eggs	0.003	0.002	
Coconut Meat	0.16	0.16	
Coconut Milk	0.08	0.24	
Coconut Crabs	0.023	0.023	
TOTAL	45.3	45.1	0.33
Island Group Janet			
Fish	44	44	
Domestic Meat	0.69	1.2	
Pandanus Fruit			1.0
Breadfruit			1.2
Wild Birds	0.10	0.020	
Bird Eggs	0.074	0.037	
Coconut Meat			2.6
Coconut Milk			3.8
TOTAL	44.9	45.2	8.6

return and for the 10-yr post-return diet. The total rate of ingestion assuming the diet at the time of return, which considers only those food items that are available at the time of return, has been used to estimate 5- and 10-yr integral dosages. The total rate of ingestion assuming the 10-yr post-return diet has been used to estimate the 30- and 70-yr integral dosages. Fish and other marine foods are seen to contribute the bulk of the plutonium to the diet. The daily rates of plutonium ingestion in Table 9 would greatly exceed that for residents of New York, whose ingestion rate of plutonium was estimated to be 1.5 pCi/yr in 1972 (BENNETT, 1974b).

Table 10. Uptake and retention parameters for ^{239}Pu

	Bone	Liver
E (MeV)	270	53
f_{man}	1.35×10^{-5}	1.2×10^{-5}
M (g)	5000	1800
λ_{man} (day^{-1})	1.91×10^{-5}	4.76×10^{-5}
λ_r (day^{-1})	-7.79×10^{-8}	7.79×10^{-8}

Doses were calculated assuming that the radionuclide concentrations in foods decrease with time by radioactive decay alone. Uptake and retention in human organs, organ mass and the energy absorbed in tissue per disintegration were adapted from ICRP (1959, 1972) reports or from the more recent literature (see Table 10). The integrated dose via food chain was determined from the equation below:

$$\text{Dose (rem)} = \frac{KEIf_{\text{man}}C_0}{M(\lambda_{\text{man}} - \lambda_r)} \times \left[\frac{1 - e^{-\lambda_r t}}{\lambda_r} - \frac{1 - e^{-\lambda_{\text{man}} t}}{\lambda_{\text{man}}}\right]$$

where

$$K = 5.1 \times 10^{-5} \frac{\text{dis/g/rem}}{\text{pCi/MeV/day}}$$

E = disintegration energy, MeV (includes RBE and n factors)
I = food intake, g/day
f_{man} = fraction of nuclide ingested reaching the organ of reference
C_0 = initial concentration of nuclide in food product, pCi/g
M = mass of the organ of reference, g
λ_{man} = effective elimination rate of nuclide from organ of reference, day-l
λ_r = radioactive decay constant, day^{-1}
t = time, day.

Dosages to bone and liver from ingestion of plutonium for island groups Alvin-Keith and Janet integrated over 5, 10, 30 and 70 yr were computed using the rates of ingestion of Table 9. The predicted 5- and IO-yr dosages from terrestrial foods for Alvin-Keith (Table 11) exceed those for Janet. These dosages assume the diet at the time of return. The greater dosage for Alvin-Keith can be explained by the absence of coconut and coconut crabs in the diet from Janet and by the greater concentrations of plutonium in the birds from Alvin-Keith. The 30- and 70-yr dosages for Janet exceed those for Alvin-Keith, as one would expect on the basis of greater plutonium contamination of the terrestrial environment on Janet. For both island groups marine foods can be expected to contribute far greater dosages than terrestrial foods. The 30- and 70-yr dosages from marine

Table 11. *Summary of predicted integral dosages from ingestion of plutonium*

A. Terrestrial Foods

	Integral Dose, rem							
	5 Yr		10 yr		30 yr		70 yr	
	Bone	Liver	Bone	Liver	Bone	Liver	Bone	Liver
Alvin-Keith	7.8(-5)	3.8(-5)	3.1(-4)	1.5(-4)	2.9(-3)	1.2(-3)	0.016	5.6(-3)
Janet	5.2(-5)	2.5(-5)	2.1(-4)	9.8(-5)	1.2(-2)	5.5(-3)	0.082	0.033

3. *Marine* Foods

All Groups	2.7(-3)	1.3(-3)	1.0(-2)	5.0(-3)	0.091	0.041	0.45	0.18

foods for Janet would be 5-10 times greater, the dosages from marine foods for Alvin-Keith and the 5- and IO-yr dosages from marine foods for Janet would be 30-50 times greater than the corresponding dosages from terrestrial foods.

Dosage via all Pathways

Table 12 shows the predicted 30-yr dosages from plutonium to the three major receptor organs (lung, liver and bone) via the three relevant exposure pathways. The major pathway for plutonium for living patterns involving residence and agriculture on the southern islands is the marine pathway, followed by the terrestrial and inhalation pathways. The 30-yr dosages via inhalation for Alvin-Keith are about 1 mrem or less and are therefore comparable to or less than the estimated dose commitments to a New Yorker through the year 2000 from inhalation of plutonium from world-wide fallout (BENNETT, 1974a). The 30-yr dosages to liver and bone from terrestrial foods for the southern islands are relatively low, approx l-3 mrem, but they exceed the 30-yr dose commitments to a New Yorker via ingestion by 2 to 3 orders-of-magnitude (BENNETT, 1974b). The dosages via marine foods, which contribute most to the total dosages are greater still. For living patterns involving residence and agriculture on Janet and other northern islands, the major pathway for plutonium is the inhalation pathway followed by the marine and terrestrial pathways. The 30-yr dosages via inhalation exceed those for Alvin-Keith by more than 2 orders-of-magnitude. The 30-yr dosages via terrestrial foods exceed those for Alvin-Keith by about a factor of five.

Table 12. *The plutonium* 30-yr *integral dose to bone, liver and lung via the three exposure pathways*

Plutonium 30-Year Integral Dose, Rem

UNMODIFIED CONDITIONS

Living	Marine			Terrestrial			Inhalation			Total		
Pattern	Bone	Liver	Lung	Bone	Liver	Lung	Bone	Liver	Lung	Bone	Liver	Lung
I	0.091	0.041	-	2.9(-3)	1.2(-3)	-	7(-4)	4(-4)	9(-4)	0.095	0.043	9(-4)
II	0.091	0.041	-	1.2(-2)	5.5(-3)	-	0.10	0.056	0.13	0.20	0.15	0.13

Living Pattern	Village Island	Agriculture	Visitation
I	Enewetak-Parry	Alvin-Keith	Southern Is.
II	Janet	Janet	Northern Is.

Tabk 13. Major contributions of radionuclides to the 30-yr integral dose to bone and liver

30-yr Integral Dose, Rem

Living Pattern I[a]

Nuclide	Marine Bone	Marine Liver	Terrestrial Bone	Terrestrial Liver	Inhalation Bone	Inhalation Liver
^{60}Co	0.017	0.18	6.2(-4)	6.6(-3)		
^{90}Sr	0.77	4.8(-4)	2.2	1.4(-3)		
^{137}Cs	0.030	0.030	0.18	0.18		
^{239,240}Pu	0.091	0.041	2.9(-3)	1.2(-3)	7(-4)	4(-4)
Total	0.91	0.25	2.4	0.19	7(-4)	4(-4)

.Total dose to bone all pathways[b] = 4.1 rem
Total dose to liver all pathways[b] = 1.3 rem

[a]See Table 12 for a description of the living patterns.
[b]The total dose via all pathways includes 30-yr external gamma doses of 0.83 rem for Alvin-Keith and 4.0 rem for Janet.

Living Pattern II[a]

Nuclide	Marine Bone	Marine Liver	Terrestrial Bone	Terrestrial Liver	Inhalation Bone	Inhalation Liver
^{60}Co	0.017	0.18	3.0(-4)	0.032		
^{90}Sr	0.77	4.8(-4)	7 4	0.046		
^{137}Cs	0.030	0.030	8.0	0.0		
239,240,	0.091	0.040	0.012	5.5(-3)	0.10	0.056
Total	0.91	0.25	02	8.1	0.10	0.056

Total dose to bone all pathways[b] = 87 rem
Total dose to liver all pathways[b] = 12 rem

[a]See Table 12 for a description of the living patterns.
[b]The total dose via all pathways includes 30-yr external gamma doses of 0.83 rem for Alvin-Keith and 4.0 rem for Janet.

Although plutonium would contribute more than 95 % of the total dosages via inhalation (NVO, 1973), this pathway is not the greatest contributor to the total dosage via all pathways (Table 13). For most living patterns, the relative ranking of the pathways from the standpoint of their contributions to the total dose would be (1) terrestrial food chain, (2) external gamma dose, (3) marine food chain, (4) inhalation. For the pathways ranking above inhalation nuclides other than plutonium would contribute most to the dosages. Thus ^{90}Sr and ^{137}Cs would contribute most via the terrestrial food chain, while ^{60}Co would make a smaller contribution, comparable to that of ^{239,240}Pu. For the marine food chain ^{90}Sr would contribute most to the bone dose and ^{60}Co, ^{137}Cs and ^{239,240}Pu would make more or less comparable contributions to the liver dose. Essentially the entire external gamma dose can be attributed to the presence of ^{137}Cs and ^{60}Co. The potential dosage from plutonium via all pathways is low relative to that from ^{90}Sr, ^{137}Cs and ^{60}Co. Remedial measures that may be designed to limit the dose via the terrestrial food chain or the external dose commitment will have very little impact on the plutonium dose via the marine food and inhalation pathways (NVO, 19 73). It must be remembered that by virtue of its very long half-life, plutonium will still remain in the Atoll environment after the other major radionuclides have decayed away.

REFERENCES

AARKROG A., 1971, *Health Phys.* **20,** 31.
BENNETT B. G., 1974a, USAEC Rept. HASL-278, Pt. 1, p. 41 (New York: USAEC HASL).
BENNETT B. G., 1974b, USAEC Rept. HASL-286, Pt. 1, p. 36 (New York: USAEC HASL).

International Commission on Radiological Protection (*ICRP*) *Publication* 2, 1959 (Oxford: Pergamon Press).

International Commission on Radiological Protection (*ICRP*) *Publication* 19, 1972 (Oxford: Pergamon Press).

National Air Pollution Control Administration (NAPCA), 1968, USHEW Rept. APTD 68-9 (Arlington, Va: NAPC).

USAEC Nevada Operations Office (NVO), 1973, Enewetak Radiological Survey, USAEC Rept. NVO-140, Vol. I, II, III (Las Vegas: USAEC NVO).

DISCUSSION

STANNARD, J. N.: Would you agree with the proposition that, if there are to be unpleasant surprises, that is, exceptions to the general rule that plutonium is poorly transportable in the environment, this exception would appear in the marine environment?

FOLSOM, T. R.: There are some data coming up to answer this question. The turnover rates of the upper layers of the ocean are being studied by physical oceanographers, and the best tool so far is ^{137}Cs. It is turned over with a half-time of about 10 yr. Plutonium is taken down by the biosphere apparently three times as fast. The ocean is still very large, very dilute, and will hold quite a lot of plutonium, which doesn't go up the chain very far-it stops at plants.

STANNARD, J. N.: I was speaking of a comparison to movement in the terrestrial environment.

WILSON, D. W.: Unpleasant surprises, if any, might occur in time as the effects of slow processes which result in enhanced biological availability become observable. These might involve shifts from inorganic plutonium to organically bound plutonium brought about by cycling of plutonium through biological systems. Estuaries, marshes, highly adapted plants in poor soils, would be the systems in which such mechanisms might occur.